Transpacific satellite and aircraft observations of Asian pollution

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Abstract

Satellite observations of carbon monoxide (CO) from the MOPITT instrument are combined with aircraft measurements from the TRACE-P aircraft mission over the northwest Pacific, and with a global 3-D model of atmospheric chemistry (GEOS-CHEM), to quantify Asian pollution outflow over the Pacific and its transpacific transport during spring 2001. Global CO column distributions in MOPITT and GEOS-CHEM are highly correlated (R^2=0.87) with no significant model bias. The model CO column over southeast Asia is overestimated by 18% relative to MOPITT, implying excessive biomass burning emissions (50-60% reduction necessary). This result is consistent with an inverse model analysis of TRACE-P CO observations. MOPITT observed four major events of transpacific transport of Asian pollution in spring 2001; these are all simulated by the GEOS-CHEM model and are seen in the in situ observations. One of them (Feb. 26-27) was sampled as a succession of pollution layers at 15-40º N by the TRACE-P aircraft over the northeast Pacific. We show that these layers all originated from one single event (February 22) of Asian outflow to the Pacific along a warm conveyor belt ahead of a cold front. This Asian outflow split into northern and southern plumes upon encountering a blocking high over the mid-Pacific (February 25). The northern branch (sampled on February 26 off of California) was not associated with elevated ozone. The southern subsiding branch (sampled on February 27) contained a 12 ppbv ozone enhancement driven by decomposition of peroxyacetylnitrate (PAN) to nitrogen oxide radicals as deduced from observations.

1. Introduction

Quantifying the continental outflow and intercontinental transport of air pollutants is one of the major challenges of atmospheric chemistry today. An integration of satellite and aircraft observations with 3-D models is a promising new approach for addressing this challenge. We present here a first attempt at such an integration through a combined model interpretation of aircraft observations of Asian pollution outflow and transpacific transport from the Transport and Chemical Characterization and Evolution over the Pacific (TRACE-P) aircraft mission, together with satellite observations of carbon monoxide (CO) from the Measurement of Pollution in the Troposphere (MOPITT) instrument. In the process we demonstrate the value of the MOPITT observations for mapping intercontinental transport of pollution, and we present the first observationally-based analysis of the production of ozone during transpacific transport of Asian pollution.

The NASA TRACE-P mission focused on the study of Asian chemical outflow over the northwest Pacific in February-April 2001 [*Jacob et al.,* this issue]. Two aircraft intensively sampled the region from bases in Hong Kong and Japan. Transit flights from the United States sampled a major transpacific pollution transport event. The MOPITT instrument [*Drummond,* 1996; *Edwards et al*., 1999] provided global coverage of CO concentrations from a polar orbit. Validation profiles for MOPITT conducted by the TRACE-P aircraft [*Jacob et al*., this issue] set the stage for a combined analysis of the TRACE-P and MOPITT data sets. A global tropospheric model is used here to provide the necessary transfer function between the MOPITT and aircraft observations, and to place these observations in the context of our current understanding of sources and transport from Asia.

Carbon monoxide, a product of incomplete combustion, is present at concentrations of 50-500 ppbv in the troposphere. It is the principal sink of OH, the main tropospheric oxidant, and as such plays a key role in controlling the oxidizing power of the atmosphere. It has a mean atmospheric lifetime of 2 months against oxidation by OH, its main atmospheric sink, and is therefore a sensitive tracer for continental outflow and intercontinental transport of pollution [*Staudt et al.*, 2001; *Liu et al.,* this issue].

Recent studies have suggested that transpacific transport of Asian pollution has significant implications for ozone and aerosol air quality in the United States [*Jaffe et al*., 1999; *Jacob et al*., 1999; *Yienger et al*., 2000]. Model studies have shown that Asian emissions elevate the general background levels of surface O_3 over the United States [*Berntsen et al.*, 1999; *Fiore et al*., 2002], and this influence could be magnified during transpacific transport events such as those documented during the Photochemical Ozone Budget of the Eastern North Pacific Atmosphere (PHOEBEA) aircraft campaigns [*Jaffe et al*., 1999; *Jaffe et al.,* 2001; *Jaffe et al.,* 2002*; Price et al.,* this issue; *Jaeglé et al.,* this issue]. *Yienger et al.* [2000] have suggested that three to five large Asian pollution events reach the western United States every spring. Asian pollution is typically exported to the Pacific via frontal lifting in warm conveyor belts (WCBs) or convection into the free troposphere [*Bey et al.,* 2001b, *Husar et al.*, 2001; *Stohl et al*., 2001; *Liu et al.,* this issue] and can then be transported across the Pacific in 5-10 days [*Jaffe et al*., 1999; *Yienger et al*., 2000; *Jaffe et al.,* 2001; *Stohl et al*., 2002]. The evolution and characteristics of one such event will be examined here, in the larger context of the transpacific pollution transport observed by MOPITT throughout the TRACE-P mission.

2. Observations and Model

2.1 TRACE-P Observations

During the TRACE-P mission two NASA aircraft (DC-8 and P3-B) were deployed from bases in Hong Kong and Japan to intensively sample outflow from Asia. Transit flights from the United States sampled a major event of transpacific transport of Asian pollution on February 26 and 27, described in Section 4. The chemical payload of the aircraft included O_3, CO, reactive nitrogen oxides (NO_y) species, and a suite of hydrocarbon and halocarbon tracers [*Jacob et al.*, this issue]. Observations of CO were made continuously with 1 Hz frequency using the Differential Absorption of CO Measurement (DACOM) instrument [*Sachse et al*., 1987]. The measurement accuracy is approximately 2%.

Fuelberg et al. [this issue] give an overview of the meteorological conditions during TRACE-P. Asian pollution was primarily exported to the Pacific in WCBs ahead of cold fronts [*Liu et al.,* this issue]. This pollution included both anthropogenic and biomass burning source influences, reflecting seasonal biomass burning in southeast Asia [*Heald et al*., this issue], and the two influences were often mixed in the outflow [*Carmichael et al.*, this issue; *Ma et al*., this issue]. Detailed anthropogenic and biomass burning emission inventories were constructed for the TRACE-P period from bottom-up information [*Streets et al*., this issue; *Heald et al*., this issue]. These inventories were used by *Palmer et al.* [this issue] in an inverse model analysis which determined the constraints from the TRACE-P CO observations towards better quantifying CO sources from East Asia. They concluded that anthropogenic emissions from China in the *Streets*

et al. [this issue] inventory were underestimated by 30%, and that biomass burning emissions from Southeast Asia in the *Heald et al*. [this issue] inventory were too high.

Several TRACE-P studies examined the Asian outflow of ozone and of its NOy precursors. Large NO_y enhancements were observed downwind from Asia and consisted mainly of HNO_3 and PAN [*Miyazaki et al.,* this issue; *Talbot et al.,* this issue]. PAN was the dominant form of NO_y in outflow associated to WCBs [*Miyazaki et al.,* this issue]. Examination of NO_y-CO correlations by *Koike et al.* [this issue] indicated an NO_y export efficiency to the free troposphere from northeastern China of 15%, with only 0.5% of the emitted NO_x remaining as NO_x in the free tropospheric outflow. Significant ozone production in Asian outflow during TRACE-P appeared to be limited to biomass burning plumes originating from low latitudes [*Tang et al*, this issue]. Ozone production in anthropogenic plumes was generally insignificant, reflecting the weak photochemical activity at that time of year [*Pierce et al.,* this issue].

2.2 MOPITT Observations

The MOPITT instrument is an nadir IR correlation spectrometer launched aboard the NASA Earth Observing System Terra spacecraft in 1999 with the objective of continually monitoring tropospheric CO and CH_4 [*Drummond,* 1996; *Edwards et al*., 1999]. It has solar backscatter channels to measure total CO and CH_4 columns, and IR emission channels to measure CO vertical profiles; correlation spectroscopy provides 2 signals for each channel. So far only the IR emission channels have been retrieved. The satellite is deployed in a polar sun-synchronous orbit with a 10:45 local equator cross-over time. Horizontal resolution is 22 km x 22 km, and cross-track scanning achieves

approximate global coverage in 3 days. CO concentrations are reported on 7 vertical levels (surface, 850, 700, 500, 350, 250, and 150 hPa) and total column.

The MOPITT observations reported here are version 3 data [http://www.eos.ucar.edu/mopitt/], which have a 10% precision on both column and mixing ratio [*Pan et al.,* 1998; *Edwards et al*., 2003]. The concentrations at the seven vertical levels constitute the state vector ($\hat{\mathbf{x}}$) of the retrieval. Current CO retrievals are based on only 4 signals, and the 7 vertical levels reported do not represent completely independent information [*Deeter et al*., submitted]. The retrieval is ill-conditioned without the inclusion of external information in the form of an *a priori* profile ($\mathbf{x_a}$) and accompanying error covariance matrix ($\mathbf{S_a}$). The vertical profile ($\hat{\mathbf{x}}$) retrieved by the instrument is related to the true vertical profile ($\mathbf{x}$) by the averaging kernel matrix ($\mathbf{A}$), as described by *Rodgers* [2000]:

$$\hat{\mathbf{x}} = \mathbf{x}_a + \mathbf{A}(\mathbf{x} - \mathbf{x}_a) + \mathbf{G}\boldsymbol{\varepsilon} \qquad (1)$$

where $\boldsymbol{\varepsilon}$ is the measurement error and $\mathbf{G}$ is a gain matrix. The MOPITT retrieval uses an invariant *a priori* CO profile and standard error based on statistics from field measurements, and a model climatology for the highest levels [*Deeter et al*., submitted]. The averaging kernel matrix is given by:

$$\mathbf{A} = \mathbf{I} - \hat{\mathbf{S}}\,\mathbf{S}_a^{-1} \qquad (2)$$

where $\mathbf{I}$ is the identity matrix and $\hat{\mathbf{S}}$ is the retrieval error covariance matrix, different for each retrieval and supplied with the MOPITT retrieval product. The rows of $\mathbf{A}$ (averaging kernels) describe the vertical resolution of the retrieval. A sample set of averaging kernels for the North Pacific is shown in Figure 1. We see that perturbations to surface CO have no effect on the retrieval; perturbations to mid-tropospheric CO

influence all reported levels; and perturbations to upper tropospheric CO have a broad upper tropospheric signature in the retrieval. There appears to be roughly two independent pieces of information in the MOPITT retrieved profile: one for CO in the middle troposphere (300-800 hPa) and one for CO in the upper troposphere (pressure < 500 hPa) [*Deeter et al*., submitted]. We find that the global average number of degrees of freedom for the signal in the MOPITT vertical profiles for one sample day (March 15, 2001) is 1.21 ± 0.45 according to the *Rodgers* [2000] definition. We focus therefore on the column data in our analysis.

Seven MOPITT validation profiles were conducted during TRACE-P for a range of conditions. Four were conducted under sufficiently clear-sky conditions to enable MOPITT retrievals. Results presented by *Jacob et al*. [this issue] show that the MOPITT column observations captured the variability in the aircraft observations, with a high bias of 6±2%. They confirm that MOPITT does not provide significant information on the vertical structure of CO. One of the validation profiles (DC-8 flight 5) sampled a substantial transpacific transport event, discussed in Section 3.

2.3 Model Description

We use the GEOS-CHEM v. 4.33 global 3-D model of tropospheric chemistry (http://www-as.harvard.edu/chemistry/trop/geos) with 2°x2.5° horizontal resolution and 48 vertical levels. The model is driven by assimilated meteorology from the Goddard Earth Observing System (GEOS)-3 of the NASA Data Assimilation Office and was initially described in *Bey et al.* [2001a]. The GEOS-3 data have a temporal resolution of 6 hours (3 hours for surface variables and mixing depths). We conducted a simulation for

February-April 2001 with full O_3-NO_x-hydrocarbon chemistry. We also conducted CO-only simulations with tagged sources and monthly mean OH concentration fields archived from the full-chemistry simulation. Initial conditions were taken from a one-year spin-up simulation and model output was sampled every 3 hours. Sources of CO and its precursor hydrocarbons are as described by *Duncan et al.* [2003]. Sources of NO_x for the full-chemistry simulation are as described by *Martin et al.* [2002]. Biomass burning emissions are from a bottom-up inventory constrained with daily satellite fire counts for the TRACE-P period [*Heald et al*., this issue]. Most of the global biomass burning for this period takes place in southeast Asia and the corresponding regional source for February-April 2001 is 69 Tg CO.

Our simulation of the ensemble of the TRACE-P CO observations is generally unbiased (mean model underestimate of 4.6 ppbv) with greatest mean discrepancies (5-15 ppbv) in the boundary layer. Other TRACE-P studies using the GEOS-CHEM model demonstrate the ability of the model to simulate the meteorological and chemical signatures of Asian outflow over the Pacific [*Kiley et al.,* this issue; *Li et al*., this issue; *Liu et al*., this issue; *Jaeglé et al*., this issue; *Palmer et al*., this issue]. Our anthropogenic source of CO from China (181 Tg CO yr^{-1}) [*Duncan et al*., 2003] is about 66% higher than in the inventory of *Streets et al.* [this issue]. An inverse model analysis by *Palmer et al.* [this issue] using the *Streets et al.* [this issue] inventory as *a priori* and GOES-CHEM as the forward model, concluded that the *Streets et al.* [this issue] inventory is too low by 30%. Their *a posteriori* estimate of anthropogenic CO emissions from China (142 Tg CO yr^{-1}) is 22% lower than ours. Their resulting simulation of the TRACE-P CO observations shows a low bias of 15 ppbv.

Comparisons between full-year model results for 2001 and ground station CO measurements by the National Ocean and Atmospheric Administration (NOAA) Climate Monitoring and Diagnostics Laboratory (CMDL) Carbon Cycle Group for representative northern hemispheric sites are shown in Figure 2. Representation error between the weekly CMDL sampling and the model monthly mean concentrations may bias these comparisons. Several recent changes to the GEOS-CHEM chemical mechanism, including a faster $N_2 + O(^1D)$ rate constant [*Ravishankara et al*., 2002] have decreased the global mean OH concentration by 10-15%. The most recent global model evaluation of CO by *Duncan et al.* [submitted], which does not include this change in rate constant, shows a model underestimate in the northern tropics and mid-latitudes by 10-40 ppbv with better agreement achieved in the arctic latitudes. The model simulation reported here is generally unbiased in its simulation of the CMDL northern hemispheric sites; however high-latitude sites are underestimated by 10-30 ppbv in summer-fall. Our 10-30 ppbv spring underestimate at Midway (MID) may be affected by the representation error mentioned above, since both the TRACE-P observations and the MOPITT observations (discussed below) do not reveal such a bias.

3. Interpretation of MOPITT observations for the TRACE-P period

Figure 3 shows the mean CO column observed by MOPITT during TRACE-P (February 20-April 10, 2001) and the corresponding GEOS-CHEM model column with MOPITT averaging kernels applied (according to equation 1). Here the MOPITT data for each day have been averaged over the 2°x2.5° GEOS-CHEM model grid and the resulting gridded product is then averaged over the 49-day TRACE-P period. MOPITT

retrievals are not performed for cloudy scenes, removing 75-80% of the total global data [personal communication, Juying Warner], but the effective coverage is increased here because of averaging over the 2°x2.5° grid. Temporal coverage of the gridded MOPITT product over the North Pacific is 20-75% (Figure 4). The GEOS-CHEM model results in Figure 3 were constructed by sampling the model on the MOPITT pressure levels for each retrieval scene, weighting by the MOPITT averaging kernels, and integrating the column following the MOPITT procedure (http://www.eos.ucar.edu/mopitt/data).

Both model and satellite observations show high CO levels over continental source and outflow regions, as well as a general latitudinal gradient. A difference plot (Figure 3, bottom panel) shows no mean global bias between the model and measured CO. Mean MOPITT and GEOS-CHEM columns for the TRACE-P period are highly correlated (R^2= 0.97, slope=0.97); and the correlation also holds for data points on individual days (R^2=0.87, slope=0.95; Figure 5). The slope indicates that the model underestimates MOPITT observations by 5%, although as pointed out in Section 2.2, the MOPITT observations appear to be biased high by 6% relative to the TRACE-P aircraft data.

The pattern of model bias relative to MOPITT matches the distribution of biomass burning [*Heald et al.,* this issue]. In particular, MOPITT column observations over Southeast Asia (10-25°N, 80-110°E) are on average 18% lower than the model column. A similar bias is found for observations over southern China, where biomass burning effluents from southeast Asia are lifted to the middle troposphere during transport in WCBs [*Liu et al*., this issue]. The discrepancy suggests that biomass burning emissions in the model for Southeast Asia may be too high. An inverse model analysis of the

TRACE-P CO data by *Palmer et al.* [this issue] arrives at the same conclusion. We find in the model that reducing the biomass burning source over Southeast Asia by about 50% would correct the discrepancy. GEOS-CHEM analysis of the TRACE-P CO data based on correlation with HCN as a tracer of biomass burning does not indicate an overestimate of biomass burning emissions [*Li et al*., this issue]. However, the use of HCN as a biomass burning tracer in Asian outflow may be compromised by a significant urban source in China [*Singh et al*., this issue]. As pointed out above, biomass burning influences were generally mixed with urban influences in the Asian outflow.

An important caveat in the interpretation of model vs. MOPITT discrepancies in terms of errors on CO sources is the possibility of model bias in the vertical structure of CO. This could arise, in particular, as a result of overly-vigorous convection transporting CO to the upper troposphere where MOPITT is most sensitive. Comparison of GEOS-CHEM and MOPITT vertical profiles over Southeast Asia shows no significant difference in shape (Figure 6), but this reflects the lack of MOPITT information on vertical structure as previously discussed. The model vertical profiles over Southeast Asia show an enhancement in the upper troposphere (~100 ppbv CO) due to deep convection (Figure 6). If we change the shape of the vertical profile in the model, while conserving the total CO column, such that we reduce upper tropospheric CO to tropical background concentrations (55-70 ppbv) and correspondingly increase lower tropospheric CO, we can match the MOPITT observations (Figure 6). Cloud data show that frequent deep convection did occur over southeast Asia during TRACE-P [*Fuelberg et al*., this issue], and an intercomparison of model simulations of CO for the TRACE-P

period reveals no systematic bias in GEOS-CHEM [*Kiley et al,* this issue], but in the absence of measured vertical profiles over southeast Asia some ambiguity remains.

The MOPITT observations reveal no evident model bias in anthropogenic emissions from China. There is also no significant model bias in the simulation of the TRACE-P aircraft data, as discussed in Section 2.3. Previous inverse model studies by *Kasibhatla et al.* [2002] and *Pétron et al*. [2003] required high anthropogenic emissions from Eastern Asia (350-380 Tg yr^{-1} and >500 Tg yr^{-1}, respectively, compared to our 323 Tg yr^{-1}) to achieve agreement with global CMDL ground station observations in 1994 and 1990-1996 respectively. It is unlikely that the emissions derived by *Pétron et al*. [2003] would be consistent with the MOPITT observations.

We now turn to the variability of Asian outflow to the Pacific and subsequent transpacific transport, as observed by MOPITT and simulated by GEOS-CHEM. Figure 7 shows MOPITT and GEOS-CHEM time series for the TRACE-P period over the northwest, central and northeast Pacific. As before, the GEOS-CHEM model results are sampled for each MOPITT scene and have the MOPITT averaging kernels applied. The MOPITT observations over the northwest Pacific show outflow events every 5-8 days associated with the passage of cold fronts across the Asian Pacific Rim, as also observed during TRACE-P [*Fuelberg et al.,* this issue; *Liu et al.,* this issue]. GEOS-CHEM and MOPITT are well correlated (R^2=0.61) in this region, reflecting the ability of the model to capture the WCBs driving the Asian outflow ahead of the frontal passages [*Liu et al.,* this issue]. Further downwind across the Pacific the agreement between model and observations degrades (R^2 over the northeast Pacific = 0.35), presumably due to accumulation of model transport errors during transpacific transport. MOPITT identifies

four major events of transpacific transport of Asian pollution during the TRACE-P period, and these are indicated by arrows in Figure 7. These events are all captured by the GEOS-CHEM model, but with a dampened magnitude.

In situ observations support the identification by MOPITT of four transpacific transport events during the TRACE-P period. Event 1 was observed on the outbound TRACE-P transit flights, as discussed further in Section 4. Events 2 and 3 were observed at the Cheeka Peak Observatory (CPO) in Washington on March 11 and 20 respectively [*Jaeglé et al*., this issue]. There were no observations at CPO during events 1 and 4. Event 4 was observed from aircraft during the PHOBEA II experiment and was characterized as an air mass which had descended from high latitudes but originated from East Asian sources [*Jaffe et al*. 2002].

Crawford et al. [this issue] showed that high CO concentrations observed over the northwest Pacific during TRACE-P were correlated with cloud cover, reflecting the role of WCBs as the dominant pathway of outflow; they suggested that this may lead to bias in satellite observations of continental outflow. To investigate this issue we compared the average CO column from GEOS-CHEM over the northwest Pacific during the TRACE-P period ($2.62 \pm 0.11 \times 10^{18}$ molecules cm^{-2}) to the average of the subset corresponding to MOPITT observation periods ($2.57 \pm 0.16 \times 10^{18}$ molecules cm^{-2}). There is no significant difference between these two numbers, suggesting that there is no cloud bias in the MOPITT observations over the northwest Pacific.

4. Transpacific Transport Event observed in TRACE-P

Layers with elevated CO from rapid transpacific transport of Asian pollution were observed on the two outbound transit flights of the DC-8 during TRACE-P (flight tracks shown in Figure 8). In flight 4 from California to Hawaii (February 26, 2001), vertical profiles indicated CO concentrations in excess of 200 ppbv in mid-tropospheric layers at 600-250 hPa (Figure 9, profiles A, B, and C; locations shown in Figure 8). The southerly layers (profiles B and C) were also enhanced in O_3, whereas the northernmost layer (profile A) at 40ºN off the coast of California was not. In flight 5 from Hawaii to Guam conducted the following day (February 27, 2001), strong CO and O_3 enhancement layers were observed in the lower free troposphere at 800-600 hPa (Figure 9, profiles D and E). CO concentrations were greater than 200 ppbv, and were accompanied by O_3 in excess of 70 ppbv.

We find that the pollution layers observed in flights 4 and 5 all originated from one single Asian outflow plume on February 22, 2001 associated with a WCB lifting Chinese pollution ahead of a cold front. Back-trajectories (Figure 8) offer evidence for this common origin; further evidence from chemical tracer observations and the GEOS-CHEM model simulation will be presented below. The Asian plume was rapidly transported across the northwest Pacific and encountered a blocking high pressure system near Hawaii on February 25 (Figure 10). It then split into northerly and southerly branches. The northern branch continued to North America and was sampled on flight 4 (February 26); the southern branch veered south to the tropical Pacific, subsided and was sampled on flight 5 (February 27). The back-trajectories for profiles B and C suggest that this Asian plume was mixed with air masses originating west of Asia or in the marine

boundary layer at these locations; we therefore limit the quantitative comparisons which follow to the observations in profiles A, D, and E.

Both the northern and southern plumes sampled in TRACE-P exhibit the characteristics of recently emitted pollution, as indicated in Table 1. The mean C_2H_2:CO concentration ratios were 2.7-3.6 pptv/ppbv, characteristic of fresh Asian outflow [*Russo et al*., this issue]. The C_3H_8:C_2H_6 ratio for the northern plume is also characteristic of fresh pollution [*Russo et al*., this issue], but the value for the southern plume is lower and indicative of some photochemical aging.

Blake et al. [this issue] examined the chemical tracer signatures for the high-CO layers in flight 5 (ascending branches only of profiles D and E). They find correlated enhancements of CH_3Cl, ethyne and halocarbons, implying a mixture of fossil fuel, biomass burning, and biofuel sources. They suggest that lower levels of halocarbons in the ascent of profile E vs. the ascent of profile D are indicative of somewhat different origins. Perchloroethene (C_2Cl_4), a chemical used in dry cleaning, is a tracer of urban pollution whereas elevated concentrations of methyl chloride (CH_3Cl) are indicative of biomass burning sources [*Blake et al.,* this issue]. The ratios for CH_3Cl/CO were similar in profiles A, D and E, and the C_2Cl_4/CO ratio was consistent within a factor of 2 (Table 1). We find that the C_2Cl_4 and CH_3Cl signatures as well as the back-trajectories (Figure 8) support a common origin not only for the elevated CO observed in profiles D and E but also for 1. Attribution of CH_3Cl to a biomass burning vs. biofuel source is ambiguous; a more specific tracer of biomass burning is the CH_3CN/HCN enhancement ratio, as CH_3CN is emitted by biomass burning but has not been observed in biofuel combustion [*Li et al.,* this issue; *Singh et al.,* this issue]. We find CH_3CN/HCN ratios >

0.6 in the pollution layers of flights 4 and 5 (Table 1), implying a biomass burning contribution to the Asian outflow [*Li et al.,* this issue]. *Blake et al.* [this issue] suggest that the higher C_2Cl_4/CO ratio in the ascent component of profile D implies a more urban contribution than in profile E. However, we find that the descent component of profile D (measured 84 km away from the ascent component) had a C_2Cl_4/CO ratio consistent with that in profile E. There is clearly some heterogeneity present in the plume, which is not inconsistent with WCB lifting air masses from different sources over East Asia.

The simulation of this transpacific transport event by GEOS-CHEM is shown in Figure 11. The GEOS-CHEM simulation indicates a dominant anthropogenic source (fossil fuel and biofuel) for CO in the outflow plume, with an additional 20-35% contribution from biomass burning effluents entrained in the WCB, as was frequently observed in TRACE-P [*Liu et al.,* this issue; *Ma et al.,* this issue; *Carmichael et al.,* this issue]. The MOPITT column observations for February 23-27, 2001 are also shown in Figure 11 along with the corresponding GEOS-CHEM model results with averaging kernels applied. Persistent clouds in the Pacific precluded a complete MOPITT retrieval; however, the agreement between MOPITT and modeled CO concentrations includes clear indications of the Asian outflow plume as it progresses across the Pacific. MOPITT observations indicate high levels of CO reaching North America on February 27.

Although the pollution layers sampled by the TRACE-P aircraft on flights 4 and 5 originated from the same outflow event from Asia, as confirmed by the back-trajectories and the chemical tracer signatures, they show evidence of very different photochemical evolution. We previously commented on the lower C_3H_8:C_2H_6 relationship observed in flight 5. The northern plume sampled in flight 4 exhibited no significant O_3 enhancement

(Figure 9, profile A). Conversely, the subsiding southern plume sampled on flight 5 contained large O_3 enhancements (Figure 9, profiles D and E). Differential absorption lidar (DIAL) observations of O_3 vertical cross-sections along the flight tracks on flights 4 and 5 (Figure 12) show no O_3 pollution enhancement at high latitudes (> 35°N) consistent with the in situ observations in the northern Asian plume. The DIAL observations show a pronounced high-O_3 layer between 2 and 4 km on flight 5, with levels exceeding 80 ppbv, consistent with the in situ observations of Figure 9 and demonstrating the broad horizontal extent (at least 2500 km) of this subsiding pollution layer.

The difference in O_3 enhancement between the northern and southern plumes can be related not only to latitude but also to the speciation of the NO_y family. Figure 13 shows the concentrations of NO_y species observed in the northern plume (profile A, flight 4) and in the southern plume (profiles D and E, flight 5). The total NO_y concentration in both plumes is 470-530 pptv, consistent with their common origin (NO_y is conserved). Mean NO_x levels were 18 pptv in the northern plume (4% of total NO_y), and 163 pptv in the southern plume (30% of total NO_y). Total NOy also included 13% HNO_3+nitrate aerosol and 74% PAN in the northern plume, vs. 51% HNO_3+nitrate aerosol and 14% PAN in the southern plume (Figure 13). This difference in speciation can be interpreted as due to PAN decomposition to NO_x in the subsiding southern plume, driving O_3 production following the split of the original plume on February 25. Temperature measurements in the plume suggest that the PAN lifetime drops from 7 weeks in profile A to 5-6 hours in profiles D and E. From Figure 8, O_3 in the southern plume appears to be enhanced by 40 ppbv above the local tropical background. However, considering the

mid-latitude origin of this plume, a background concentration of 56 ppbv (such as seen in profile A, Table 1) is more appropriate for quantifying O_3 production. By contrasting the O3 concentrations in profiles D and E vs. profile A in Table 1, we infer an ozone production of 12 ppbv in the southern plume in the 2 days following the splitting of the plume. Photochemical model calculations for profiles D and E in flight 5 [*Olson et al*., this issue] indicate an equivalent 24-hour net O_3 production rate of 0.1-0.2 ppbv/hr in the plume. This production can explain only half of the observed O_3 increment, suggesting that the plume had realized most of its O_3 production potential by the time it was sampled.

5. Conclusions

We have examined the outflow and transpacific transport of Asian pollution to by integrating aircraft observations from the TRACE-P mission (February-April 2001) with concurrent satellite observations of CO from the MOPITT instrument, and a global 3-D simulations with the GEOS-CHEM model. We use the CO column data from MOPITT after establishing that the instrument does not effectively provide additional information on the vertical profile. Four MOPITT validation profiles conducted by the TRACE-P aircraft indicate that MOPITT reproduces the variability in the in situ column observations and is biased high by 6%. The GEOS-CHEM model, which gives an unbiased simulation of TRACE-P observations of Asian outflow of CO over the northwest Pacific, is found to be highly correlated and globally unbiased with respect to the MOPITT observations during TRACE-P (R^2=0.87, slope=0.95). There are some

regional biases, the largest of which is an 18% model overestimate of MOPITT CO columns over Southeast Asia. A 50% decrease in seasonal biomass burning from that region would resolve this discrepancy, and such a correction would be consistent with an inverse model analysis of the TRACE-P CO observations using GEOS-CHEM [*Palmer et al*., this issue]. An important caveat is that model simulation of the MOPITT column CO is sensitive (through the instrument averaging kernel) to the shape of the CO vertical profile, for which MOPITT provides no independent information. As an extreme case, suppression of deep convection over Southeast Asia in the model would also resolve the regional discrepancy with MOPITT without any change to the biomass burning source. This result highlights the importance of in situ measurements of vertical profiles from aircraft to assist the interpretation of satellite observations.

The MOPITT observations for February-April 2001 identify four major events of transpacific transport of Asian pollution to North America. The GEOS-CHEM model reproduces these events and shows high correlation with the corresponding MOPITT data. All four events were sampled in situ over the northeast Pacific by either the TRACE-P or the PHOBEA-II aircraft missions, or by the Cheeka Peak Observatory in Washington State.

TRACE-P transit flights from California to Hawaii (February 26) and Hawaii to Guam (February 27) observed a succession of Asian pollution layers with CO approaching 300 ppbv, and O_3 approaching 80 ppbv, over a range of latitudes from 15º to 40ºN. Back-trajectories and chemical tracers reveal that these layers all originated from a single Asian outflow event on February 22 when a warm conveyor belt ahead of a cold front lifted Asian anthropogenic and biomass burning pollution to the free troposphere.

The MOPITT observations and the GEOS-CHEM model simulation allow a reconstruction of the evolution of this Asian outflow plume during transpacific transport. The plume split into southern and northern components upon encountering a blocking high pressure system in the mid-Pacific. Elevated O_3 (ΔO_3=12 ppbv) was observed in the southern but not in the northern plume. Reactive nitrogen in the northern plume was present mostly as PAN, but subsidence in the southern plume was found to drive PAN decomposition and from there O_3 formation. This represents the first field observation of PAN decomposition driving O_3 production in polluted plumes transported to the remote troposphere.

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Figure 9: Vertical concentration profiles of CO (black) and O_3 (red) measured from the DC-8 aircraft during the TRACE-P transit flight 4 from California to Hawaii and flight 5 from Hawaii to Guam on February 25 and 26, 2001. Profile locations and the flight tracks for flight 4 and flight 5 are shown in Figure 8. Profile A was a double spiral (ascending and then descending) conducted for MOPITT validation to check the consistency of the vertical structure of CO; data from both spirals are shown (there were no observations between 500 and 400 hPa during the ascent). CO measurements between 550 and 300 hPa in profile B are not available. Profiles D and E include descents and ascents separated by 86 and 275 km respectively.

Figure 10: Streamlines at 500 hPa for February 25, 1800 UTC indicating the blocking high pressure system over the eastern Pacific which led to splitting of the Asian pollution plume sampled on TRACE-P DC-8 flights 4 and 5. (NCEP reanalysis data provided by the NOAA-CIRES Climate Diagnostics Center, Boulder, Colorado, USA, from their Web site at http://www.cdc.noaa.gov/). The arrows illustrate the transport and splitting of the Asian pollution plume.

Figure 11: CO columns during the transpacific Asian pollution event on February 23-27, 2001 sampled by the TRACE-P aircraft on February 26-27. Left: MOPITT observations. Center: GEOS-CHEM model results sampled along MOPITT orbit tracks and with averaging kernel applied. Right: original GEOS-CHEM model results sampled at 12 GMT. The color bar is saturated at 3.5 $x10^{18}$ molecules/cm^2.

Figure 12: Cross-section of O_3 concentrations measured by DIAL along the tracks of TRACE-P flight 4 between California and Hawaii (left panel) and flight 5 between Hawaii and Guam (right panel). See Figure 8 for detail of flight tracks. Extensive Asian pollution layers were sampled between 5 and 10 km altitude on flight 4 and between 2 and 4 km altitude on flight 5. These layers contained elevated O_3 only south of 35ºN, as shown on the figure.

Figure 13: Concentrations of NOy species in the northern and southern Asian pollution plumes observed by the TRACE-P DC-8 aircraft on February 26, 2001 during flight 4 between California and Hawaii (profile A between 600-370 hPa) and February 27, 2001

during flight 5 between Hawaii and Guam (profiles D and E between 800-600 hPa). The concentrations are averages of 8 and 21 observations respectively.

Table 1: Average species concentrations and ratios observed in the transpacific Asian plume.

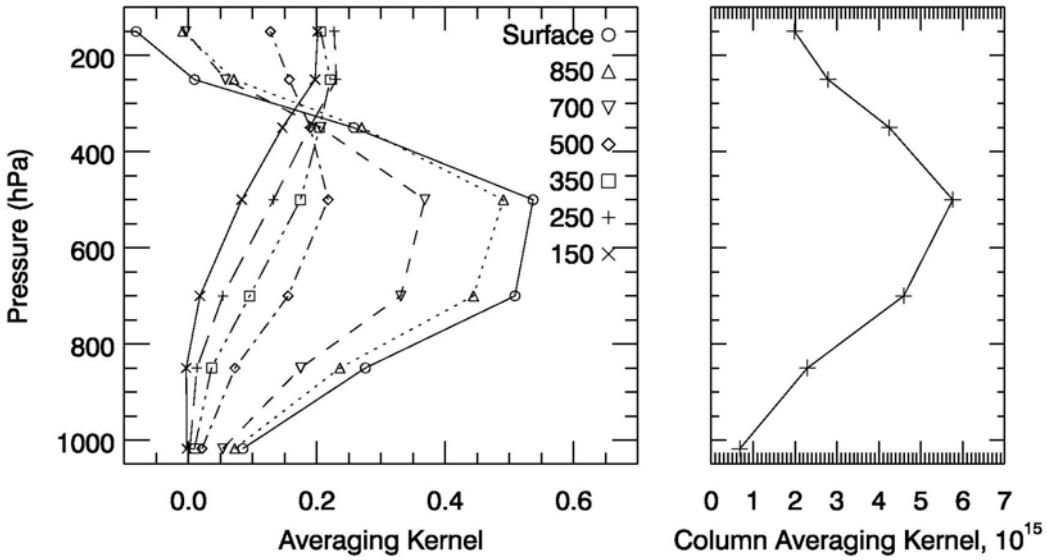
	Profile A[†,§]	**Profile D**[‡,§]	**Profile E**[‡,§]
CO (ppbv)	224 ± 28	197 ± 23	200 ± 27
O_3 (ppbv)	56 ± 2	71 ± 6	65 ± 6
C_2H_2/CO (pptv/ppbv)	3.6 ± 0.2	2.9 ± 0.3	2.7 ± 0.5
C_3H_8/C_2H_6 (pptv/pptv)	0.27 ± 0.05	0.14 ± 0.01	0.15 ± 0.02
C_2Cl_4/CO (pptv/ppbv)	0.041 ± 0.008	0.025 ± 0.013	0.038 ± 0.013
CH_3Cl/CO (pptv/ppbv)	2.8 ± 0.3	3.2 ± 0.3	3.1 ± 0.4
CH_3CN/HCN (pptv/pptv)	0.67 [*]	0.63 [*]	0.73 [*]

[†] Sampled between California and Hawaii on February 26 (average between 600-370 hPa, with standard deviation).

[‡] Sampled between Hawaii and Guam on February 27 (average between 800-600 hPa, with standard deviation).

[§] See Figure 8 for the locations of the profiles and Figure 9 for the vertical structure of the profiles.

[*] Less than 3 coincident observations, therefore no standard deviation can be defined.



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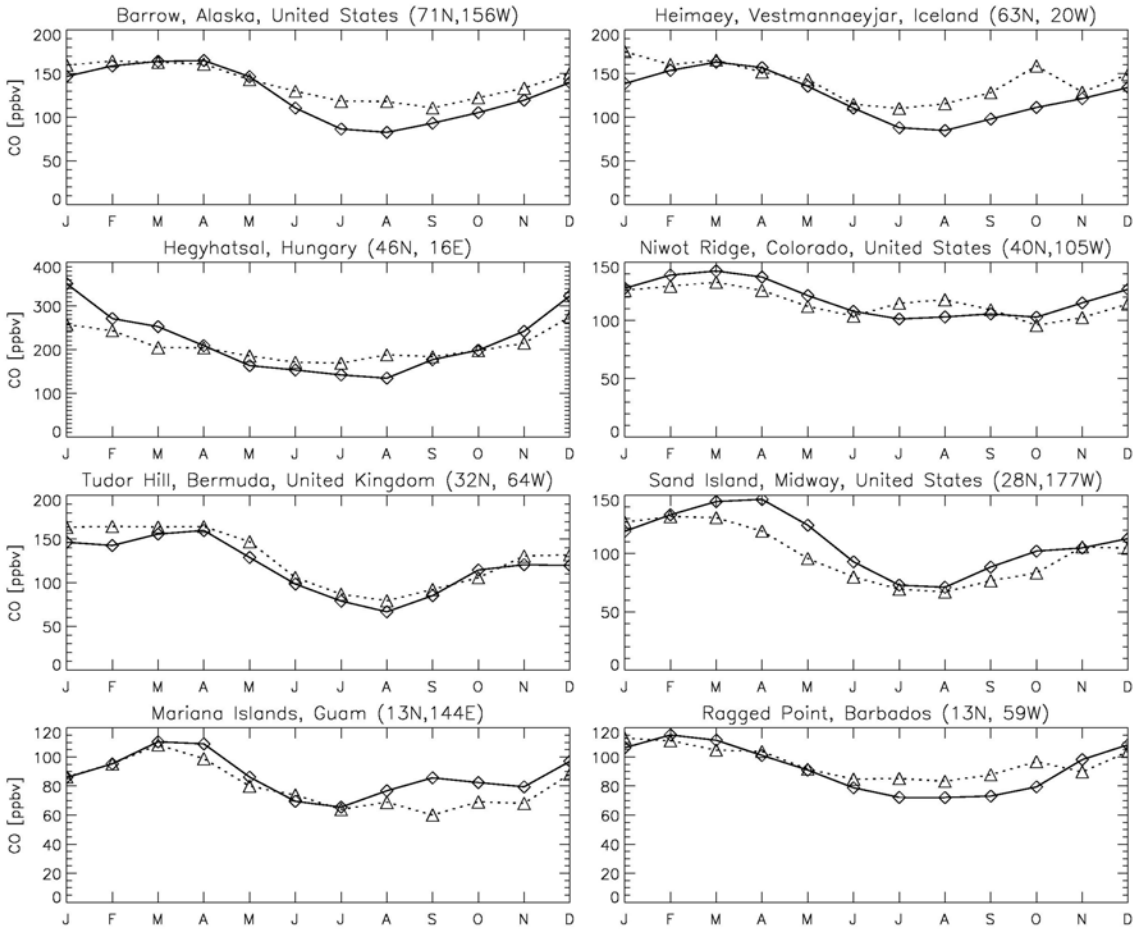

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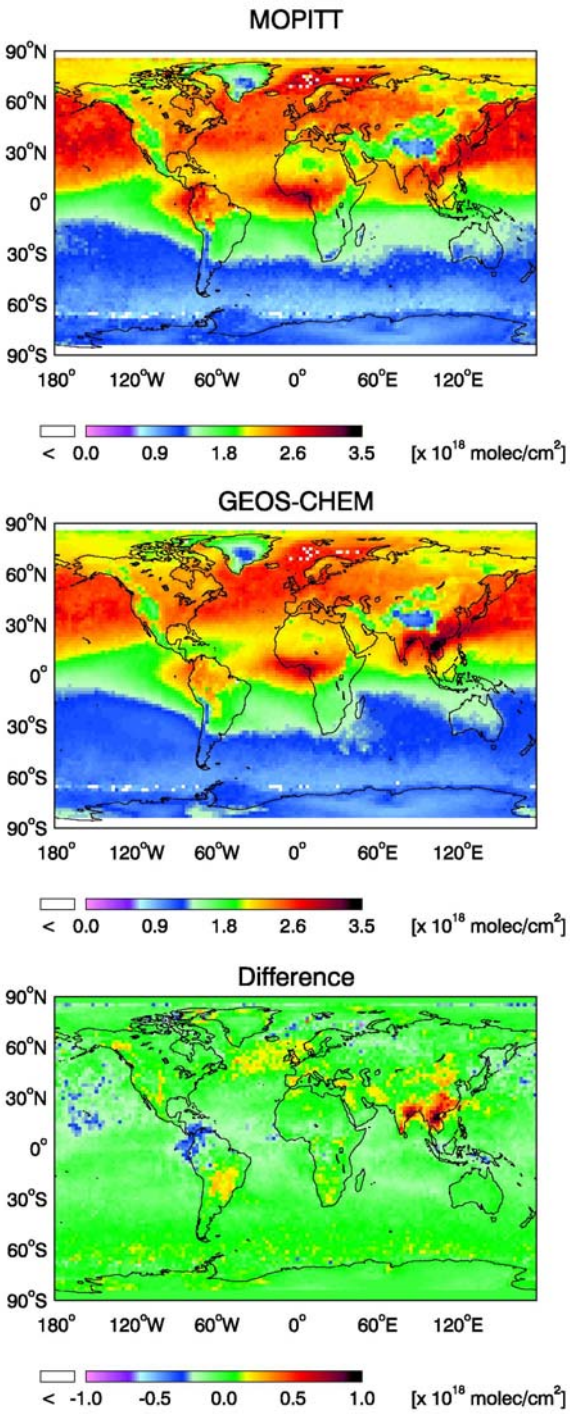

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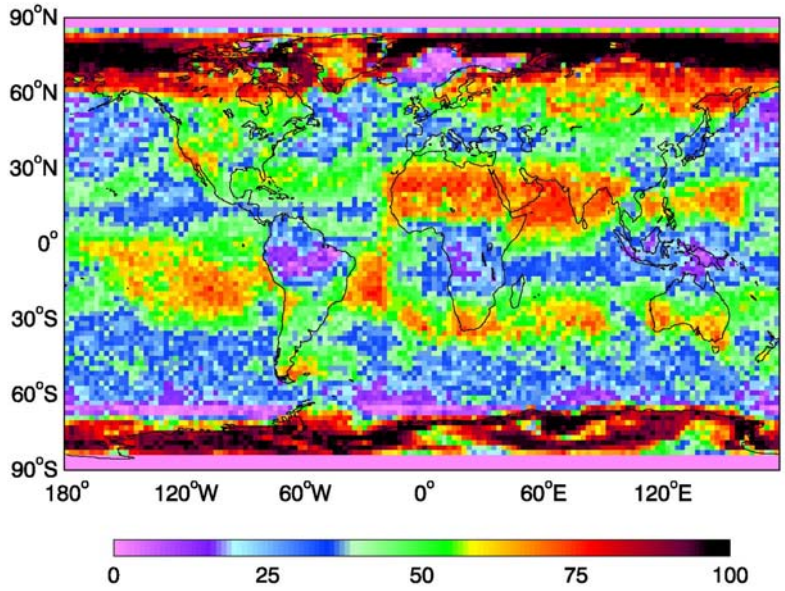

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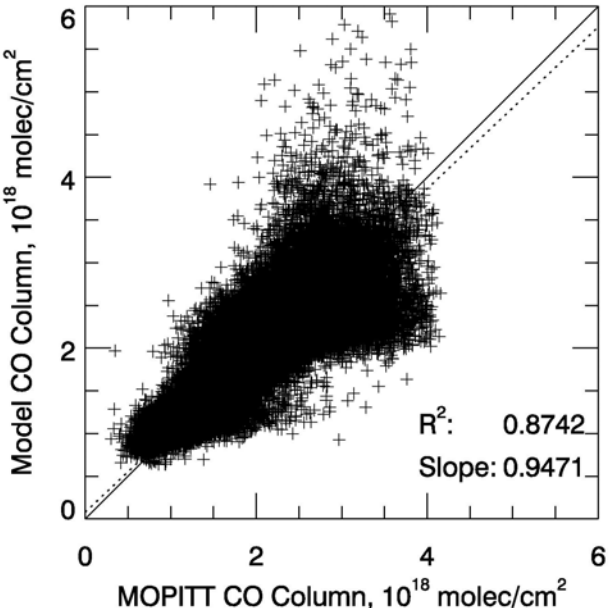

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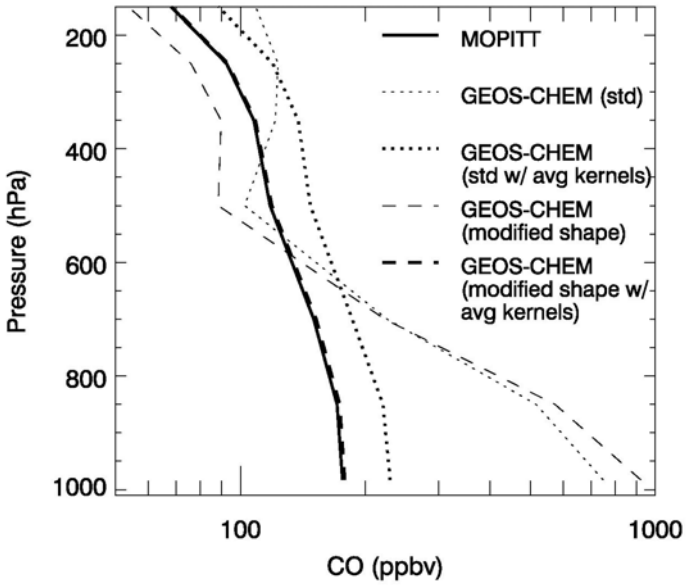

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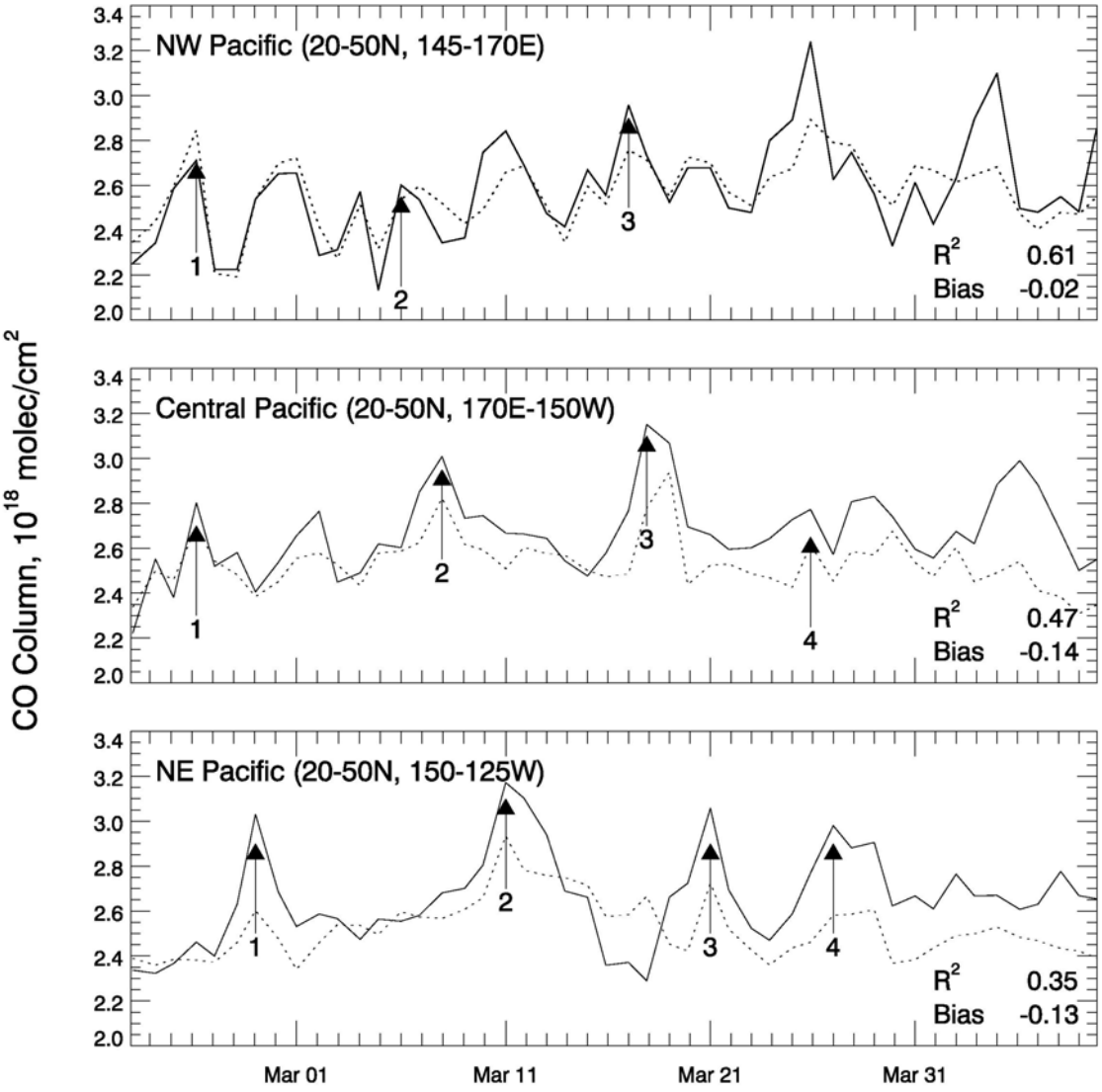

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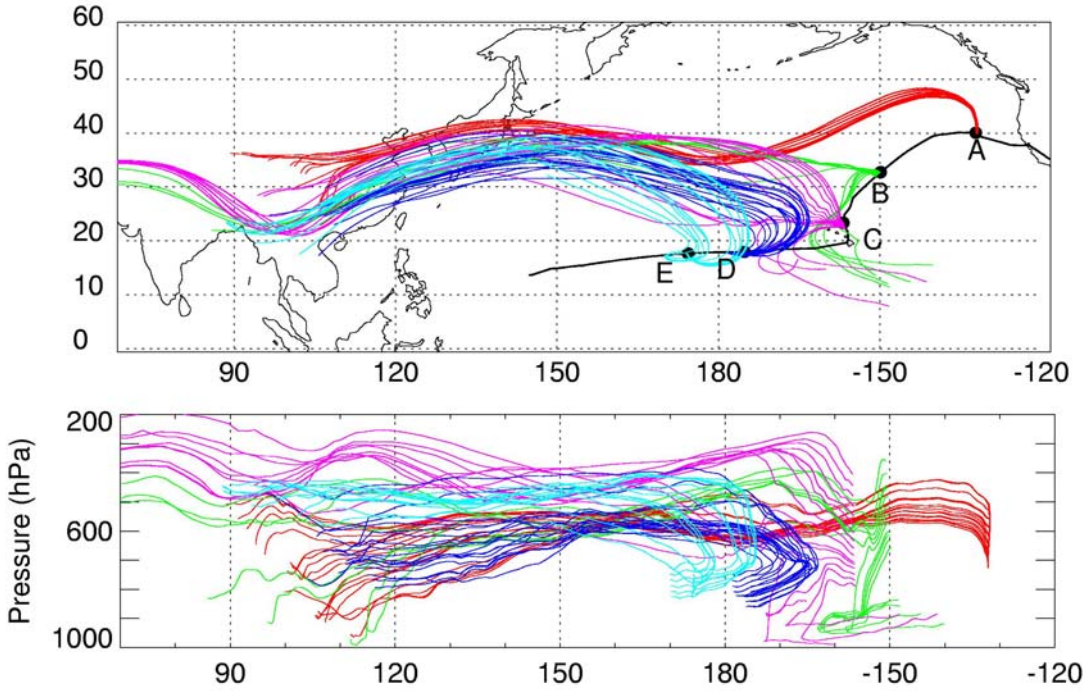

Figure 8: Five-day kinematic back trajectories based on ECMWF meteorological fields [*Fuelberg et al*., this issue] for the enhanced CO layers observed in TRACE-P DC-8 flight 4 from California to Hawaii (February 26) and flight 5 from Hawaii to Guam (February 27). The flight tracks are shown in black and arrival points in profiles A-E of Figure 9 are shown in red, green, purple, blue, and cyan respectively.

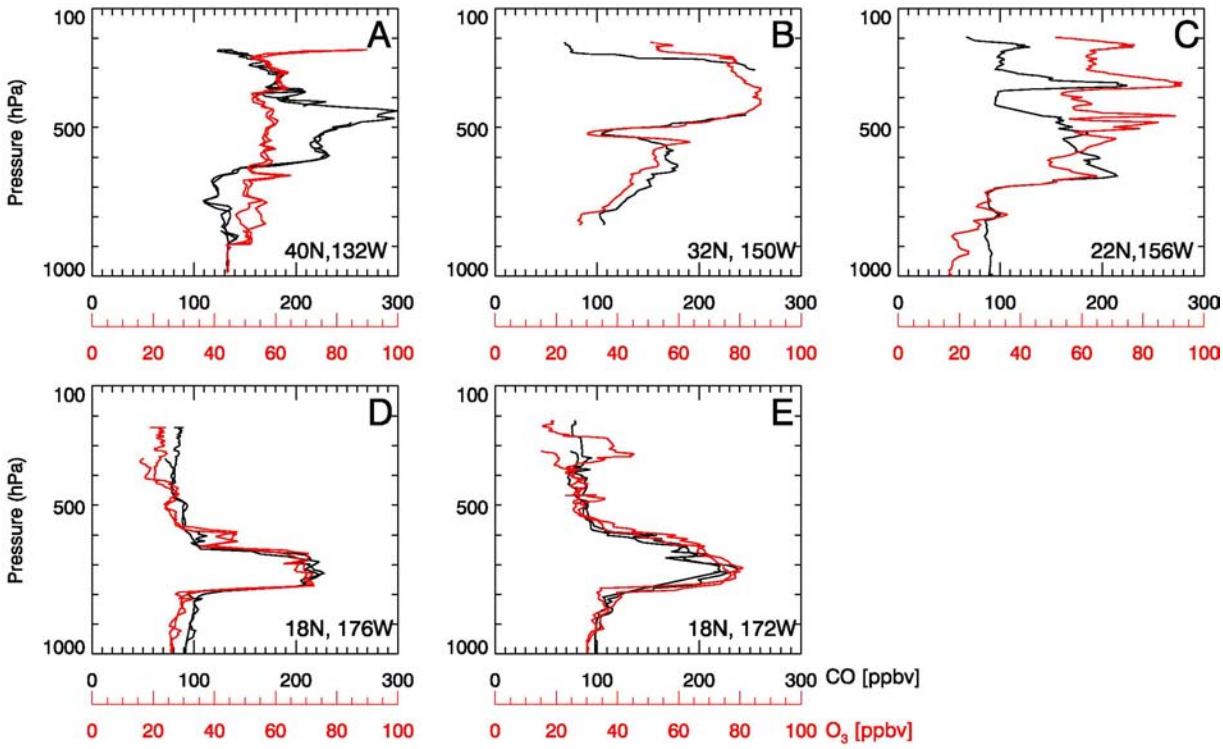

Figure 9: Vertical concentration profiles of CO (black) and O_3 (red) measured from the DC-8 aircraft during the TRACE-P transit flight 4 from California to Hawaii and flight 5 from Hawaii to Guam on February 25 and 26, 2001. Profile locations and the flight tracks for flight 4 and flight 5 are shown in Figure 8. Profile A was a double spiral (ascending and then descending) conducted for MOPITT validation to check the consistency of the vertical structure of CO; data from both spirals are shown (there were no observations between 500 and 400 hPa during the ascent). CO measurements between 550 and 300 hPa in profile B are not available. Profiles D and E include descents and ascents separated by 86 and 275 km respectively.

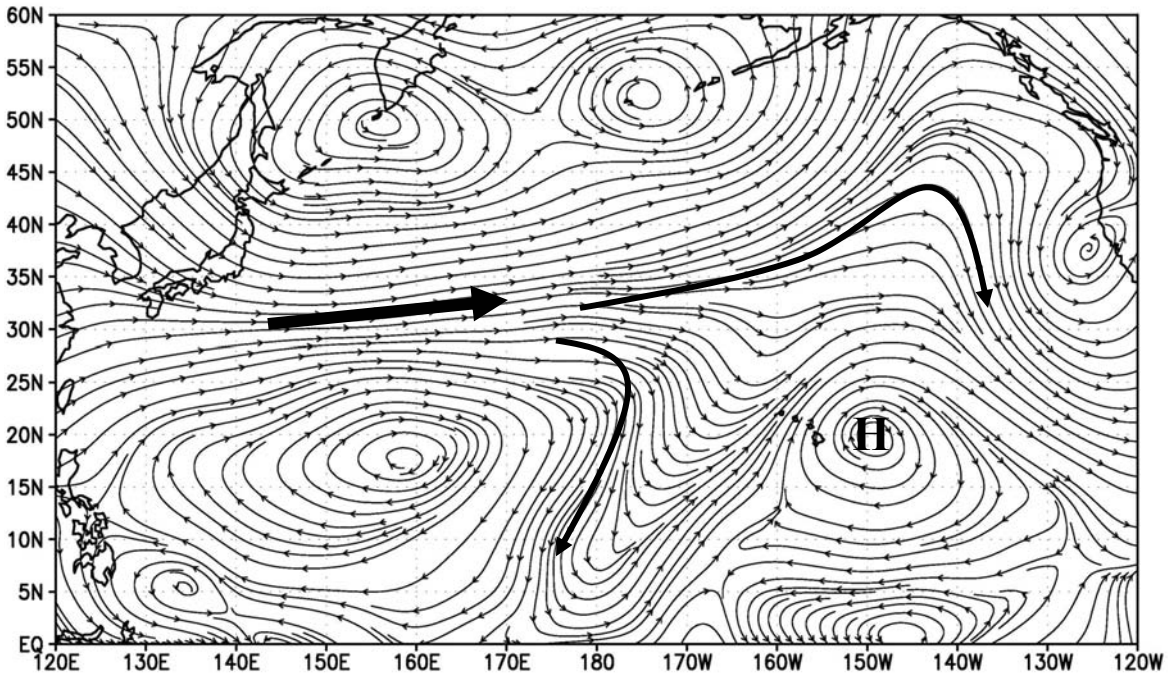

Figure 10: Streamlines at 500 hPa for February 25, 1800 UTC indicating the blocking high pressure system over the eastern Pacific which led to splitting of the Asian pollution plume sampled on TRACE-P DC-8 flights 4 and 5. (NCEP reanalysis data provided by the NOAA-CIRES Climate Diagnostics Center, Boulder, Colorado, USA, from their Web site at http://www.cdc.noaa.gov/). The arrows illustrate the transport and splitting of the Asian pollution plume.

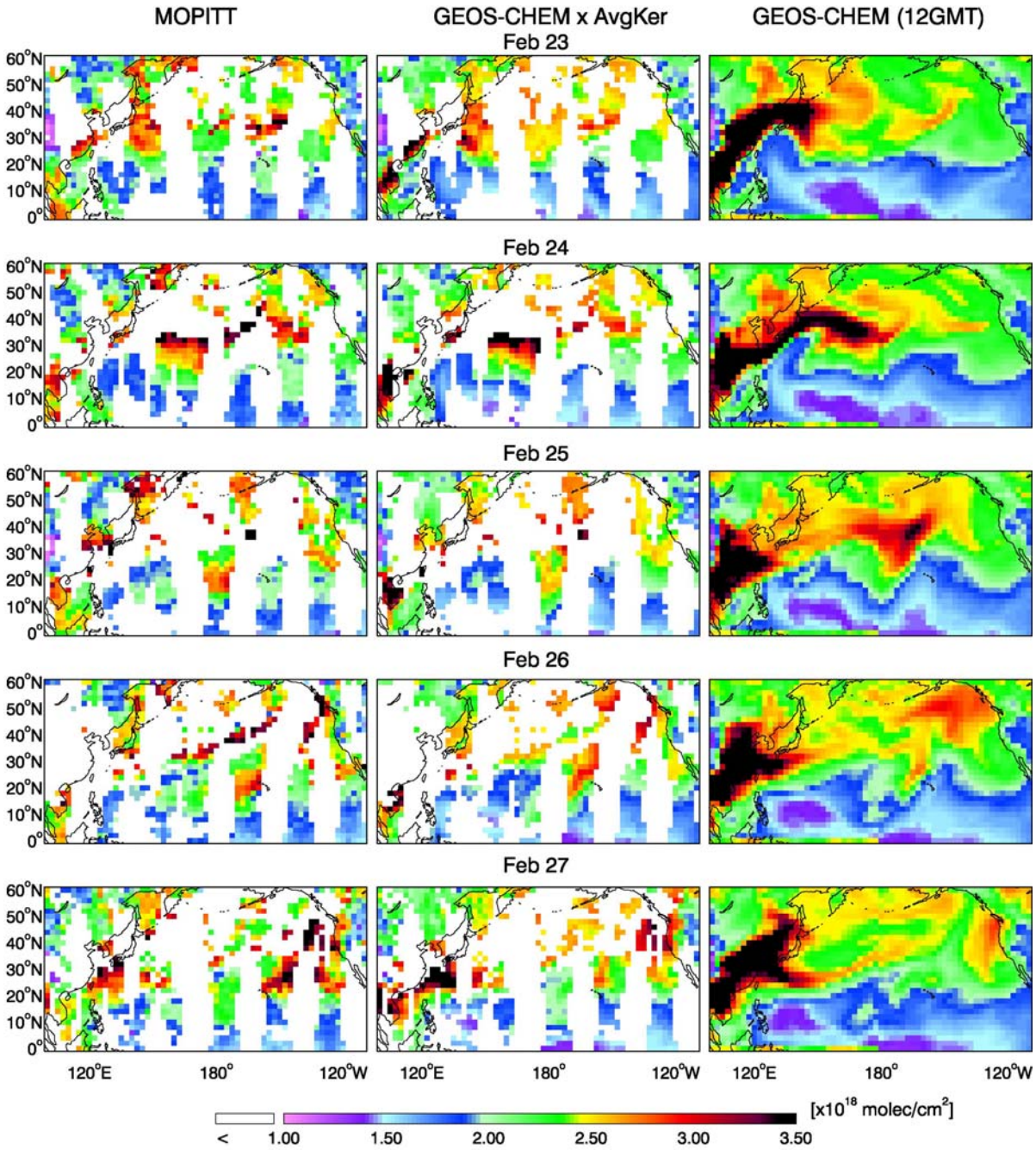

Figure 11: CO columns during the transpacific Asian pollution event on February 23-27, 2001 sampled by the TRACE-P aircraft on February 26-27. Left: MOPITT observations. Center: GEOS-CHEM model results sampled along MOPITT orbit tracks and with averaging kernel applied. Right: original GEOS-CHEM model results sampled at 12 GMT. The color bar is saturated at 3.5 x10^{18} molecules/cm^2.

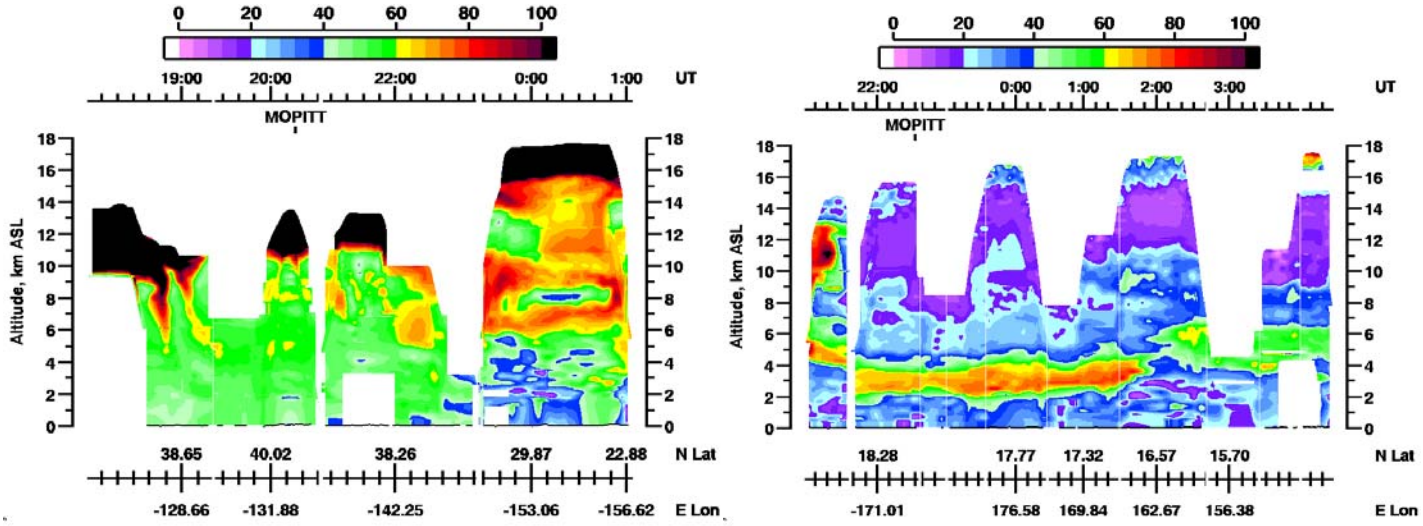

Figure 12: Cross-section of O_3 concentrations measured by DIAL along the tracks of TRACE-P flight 4 between California and Hawaii (left panel) and flight 5 between Hawaii and Guam (right panel). See Figure 8 for detail of flight tracks. Extensive Asian pollution layers were sampled between 5 and 10 km altitude on flight 4 and between 2 and 4 km altitude on flight 5. These layers contained elevated O_3 only south of 35ºN, as shown on the figure.

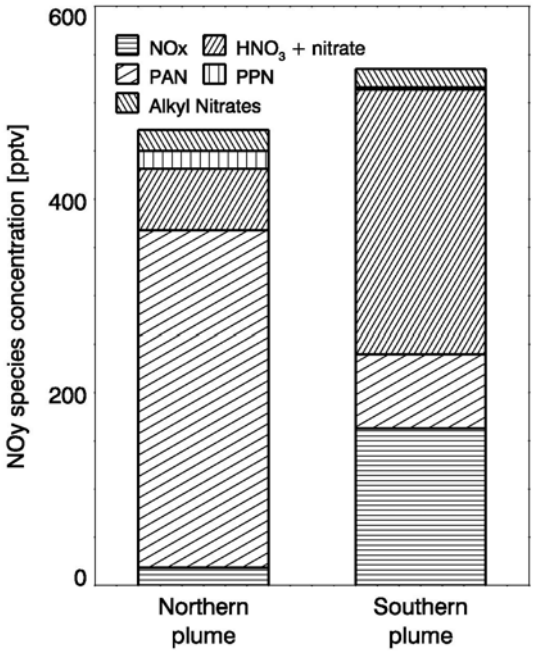

Figure 13: Concentrations of NOy species in the northern and southern Asian pollution plumes observed by the TRACE-P DC-8 aircraft on February 26, 2001 during flight 4 between California and Hawaii (profile A between 600-370 hPa) and February 27, 2001 during flight 5 between Hawaii and Guam (profiles D and E between 800-600 hPa). The concentrations are averages of 8 and 21 observations respectively.